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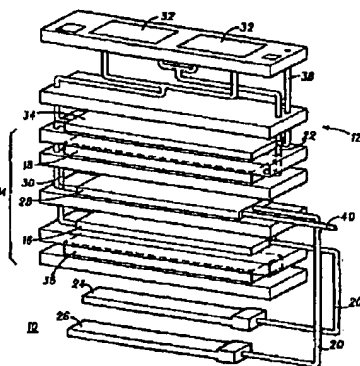
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(54) Title: FUEL PROCESSOR WITH INTEGRATED FUEL CELL UTILIZING CERAMIC TECHNOLOGY



(57) Abstract: A fuel processor (10) and integrated fuel cell including a monolithic three-dimensional multilayer ceramic carrier structure (12) defining a fuel reformer (14) and including an integrated fuel cell stack (16). The reformer includes a vaporization zone (18), a reaction zone (20) including a catalyst, and an integrated heater (22). The integrated heater is thermally coupled to the reaction zone. The fuel processor further includes a fuel channel (24) for liquid fuel and an oxidant channel (26) for hydrogen enriched gas. The fuel processor is formed utilizing multi-layer ceramic technology in which the ceramic layers are assembled from shaped to provide miniature dimensions in which the corresponding catalyst converts or reformer inlet fuel into a hydrogen enriched gas.

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FUEL PROCESSOR WITH INTEGRATED
FUEL CELL UTILIZING CERAMIC TECHNOLOGY

This application has been filed in the United States of
5 America as patent application number 09/649553 filed on
August 28, 2000

Field of Invention

10

The present invention pertains to fuel cells, and
more particularly to a fuel processor and integrated fuel
cell fabricated utilizing ceramic technology for improved
15 size and performance benefits.

Background of the Invention

20

Fuel cells in general, are "battery
replacements", and like batteries, produce electricity
through an electrochemical process without combustion.
The electrochemical process utilized provides for the
25 combining of hydrogen with oxygen from the air. The
process is accomplished utilizing a polymer electrolyte
membrane (PEM) which conducts protons sandwiched between

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two electrodes, namely an anode and a cathode. Fuel
cells, as known, can provide perpetual electricity as
long as fuel and oxygen is supplied. Hydrogen is
typically used as the fuel in fuel cells for producing
5 the electricity and it can be processed from methanol,
natural gas, petroleum, ammonia, or stored in metal
hydrides, carbon nanotubes, or as pure hydrogen.
Reformed hydrogen fuel cells (RHFCs) utilize hydrogen
fuel processed from liquid or gaseous hydrocarbon fuels,
10 such as methanol, using a fuel reformer.

Reformed hydrogen fuel cells preferably utilize
methanol that is reformed into hydrogen as a fuel source.
Methanol is the preferred fuel for use in fuel reformers
for portable applications because it is easier to reform
15 into hydrogen gas at a relatively low temperature
compared to other hydrocarbon fuels such as ethanol,
gasoline, or butane. The reforming or converting of
methanol into hydrogen usually takes place by one of
three different types of reforming. These three types
20 are steam reforming, partial oxidation reforming, and
autothermal reforming. Of these types, steam reforming
is the preferred process for methanol reforming because
it is the easiest to control and produces a higher

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hydrogen output, at a lower temperature, thus leading
itself to favored use. During steam reforming, raw
methanol is catalytically converted, with the application
of heat, to a hydrogen enriched fuel gas for use with
5 fuel cells.

Fuel reformers have been developed for use in
conjunction with fuel cell devices, but they are
cumbersome and complex devices consisting of several
discrete sections connected together with gas plumbing
10 and hardware to produce hydrogen gas, and are thus not
suitable for portable power source applications. To
date, no fuel reformers have been developed utilising
ceramic monolithic structures in which the
miniaturisation of the reformer can be achieved.
15 Laminated ceramic components, utilising ceramic
technology, are now commonly being developed for use in
microfluidic chemical processing and energy management
systems. Monolithic structures formed of these laminated
ceramic components provide for components that are inert
20 and stable to chemical reactions and capable of
tolerating high temperatures as well as providing for
miniaturized components, with a high degree of electronic
circuitry or components embedded or integrated into such

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a ceramic structure for system control and functionality.
Additionally, the ceramic materials used to form ceramic
devices including microchannels are considered to be
excellent candidates for catalyst supports in
5 microreactor devices for generating hydrogen used in
conjunction with miniaturized fuel cells.

Accordingly, it is an object of the present
invention to provide for a miniaturized fuel processor
that provides for the reforming of a fuel to a
10 hydrogen()enriched fuel gas for use in conjunction with
an integrated fuel cell.

It is yet another object of the present invention to
provide for a monolithic structure for the reforming of a
fuel to a hydrogen enriched gas.

15 It is still another object of the present invention
to provide for a monolithic structure that is formed
utilizing ceramic technology, thereby providing for the
integration of a plurality of internal plumbing
interconnections and electrical circuitry and
20 connections.

It is another object of the present invention to
provide for a fuel processor that is miniaturized for use

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in conjunction with an integrated fuel cell for portable
device applications.

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Summary of the Invention

The above problems and others are at least partially
solved and the above purposes and others are realized in
a fuel processor and integrated fuel cell including a
three-dimensional multilayer ceramic carrier structure
defining a fuel reformer and including an integrated fuel
cell stack. The fuel reformer includes a vaporisation
zone and a reaction zone including a reforming catalyst.
The ceramic carrier further includes an integrated heater
thermally coupled to the vaporization and reaction zones,
an inlet channel for liquid fuel and an outlet channel
for hydrogen enriched gas. The fuel processor is formed
utilizing ceramic technology in which thin ceramic layers
are assembled then sintered to provide miniature
dimensions in which the encapsulated catalyst converts or
reforms inlet fuel into a hydrogen enriched gas.

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Brief Description of the Drawings

The novel features believed characteristic of the
5 invention are set forth in the claims. The invention
itself, however, as well as other features and advantages
thereof will be best understood by reference to detailed
descriptions which follow, when read in conjunction with
the accompanying drawings, wherein:

10 FIG. 1 is a simplified orthogonal view of a fuel
processor and integrated fuel cell stack including a
plurality of fluidic channels, according to the present
invention;

15 FIG. 2 is a simplified sectional view of a fuel
processor and integrated fuel cell stack according to the
present invention;

FIG. 3 is a simplified sectional view of an
alternative embodiment of the fuel processor and
integrated fuel cell stack of the present invention;

20 FIG. 4 is a simplified sectional view of a thermally
conductive structure according to the present invention;

FIGs. 5 and 6 are simplified sectional views of
alternative embodiments of thermally controlled

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vaporizer/reaction zone channels according to the present invention,

FIG. 7 is a simplified flow diagram of the method of reforming fuel according to the present invention, and

5 FIG. 8 is a schematic diagram of the fuel cell system integrated with a fuel reformer according to the present invention.

Description of the Preferred Embodiments

10

Turning now to the drawings, and particular FIGs. 1 and 2, illustrated in simplified orthogonal view (FIG. 1), and in simplified cross-sectional view (FIG. 2) is a fuel processor 10 including a plurality of microfluidic
15 channels, according to the present invention. Fuel processor 10 is comprised of a three-dimensional multi-layer ceramic structure 12. Ceramic structure 12 is formed utilizing multi-layer laminate ceramic technology. Structure 12 is typically formed in component parts which
20 are then sintered in such a way as to provide for a monolithic structure. Ceramic structure 12 has defined therein a fuel reformer, or fuel processor, generally referenced 14. Fuel reformer 14 includes a vaporization

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chamber, or vaporization zone, 16, a reaction chamber, or
reaction zone, 18, and an integrated heat source, 28.
Ceramic structure 12 further includes at least one fuel
inlet channel 20 in communication with a liquid fuel
5 source of a combination solution methanol/water source 24
and a pure methanol source 26 and a hydrogen enriched gas
outlet channel 22.

During operation, heat is efficiently transferred
from the central aspect of the device, more particularly
10 an integrated heater 28, to the reaction zone 18 and fuel
vaporiser, or vaporization zone 16 using thermal
conductive channels, or vias, (discussed presently). In
this particular embodiment, integrated heater 28 is
described as a chemical heater, including a catalyst and
15 arranged so as to oxidize fuel to produce heat, but it
should be understood that the integration of an
electrical heater is anticipated by this disclosure.
Chemical heater 28 includes an air port 40 for providing
oxygen for oxidation of methanol/water 24 and/or pure
20 methanol 26 and an inlet channel 20, for providing
methanol/water 24 and/or pure methanol 26 to heater 28.

Output from fuel vaporiser zone 16 travels via
channel 30 to reaction zone 18 and then through hydrogen

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enriched gas outlet channel 22 to a fuel cell stack 32 to supply hydrogen fuel to stack 32. Spent gases from the fuel cell 32 are directed through a waste heat recovery zone 34 to capture the heat from the spent gases. Spent gases from chemical heater 28 also travel through this zone for the same reason.

An efficient thermal insulator 36 is positioned under fuel vaporizer zone 16 to keep outer temperatures low for packaging and also to keep heat localized to the fuel reformer system. As illustrated in FIGs. 1 and 2, in this particular example, high temperature fuel cell 32 is integrated with fuel reformer 14. This particular fuel cell design allows for the operation of the fuel cell at a temperature ranging from 140-200°C. Fuel vaporizer zone 16 operates at a temperature ranging from 120-160°C and the steam reforming reaction zone 18 operates at a temperature ranging from 200-300°C.

Additionally, in this particular embodiment of fuel processor 10, included is an exhaust gas vent 38, for the venting of exhaust gases generated by device 10, an air inlet 40 and a top current collector or cap 42 (as illustrated in FIG. 3).

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It should be understood that alternative embodiments encompassing: (i) alternative fuel delivery means, either passive or active pumping; (ii) fuel vaporizer, reaction zone, and chemical heater positions; and (iii) a fuel reformer device without an integrated fuel cell, are anticipated by this disclosure. In particular, anticipated is an embodiment in which only a single fuel supply, namely methanol and water, is anticipated. This use of a single methanol and water solution would enable the fabrication of a simpler design, without any need for the device to incorporate two fuel tanks. Although it is understood that pure methanol is more efficient and preferred with respect to the chemical heater, a 1 mole water and 1 mole methanol solution will also work, but is not deemed literally as operationally efficient. Further, a heater using the water and methanol solution is suitable for practical applications, and would permit a simple common fuel reservoir for feeding the heater and reformer chambers. In this instance, the fuel delivery would be split into two chambers, the chemical heater 28 and fuel vaporizer 16.

Next, anticipated are variations on the actual design of device 10 and more particularly to the actual

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location of the fuel vaporiser zone 16, reaction zone 18
and chemical heater 28. In one particular alternative
embodiment, it is anticipated that reaction zone 18
surrounds the chemical heater 28 on both sides (top and
5 bottom). In yet another alternative embodiment, it is
anticipated that reaction zone 18 can be positioned below
heater 28 and the fuel vaporiser zone 16 on top of the
chemical heater 28. In addition, and as previously
stated, it is anticipated that chemical heater 28 can
10 alternatively be an electrical heater (not shown).

Finally, it is anticipated by this disclosure that
although illustrated in FIGs. 1 and 2 is the integration
of fuel cell stack 32, a design in which a fuel cell is
not integrated with reformer 14 is additionally
15 anticipated and illustrate in FIG. 3, discussed
presently. As illustrated in FIGs. 1 and 2, when fuel
cell stack 32 is integrated with fuel reformer 14,
advantage can be taken of the heat of the substrate to
operate high temperature fuel cell stack 32. For high
20 power applications, it is convenient to design a separate
fuel cell stack and a fuel processor unit and couple them
to supply the fuel for the fuel cell. In such instances,
when a fuel cell stack is not integrated with the fuel

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reformer, and the fuel reformer is designed as a stand
alone device as illustrated in FIG. 3, external
connection can be made to connect the stand alone fuel
reformer to a traditional fuel cell stack for higher
5 power applications.

Referring now to FIG. 3, illustrated is a stand
alone fuel reformer 10'. It should be noted that all
components similar to the components illustrated in FIGs.
1 and 2 are designated with similar numbers, having a
10 prime added to indicate the different embodiment. As
illustrated, reformer 10' includes a combination fuel
comprised of liquid methanol and water, input through
inlet 10', vaporizer 16', electrical heaters 28', reaction
zone 18', and hydrogen enriched fuel outlet 22'. Device
15 10' operates generally similar to device 10 as illustrated
in FIGs. 1 and 2, except in this particular embodiment,
and hydrogen enriched fuel outlet 22' serves to direct the
reformed hydrogen fuel toward an externally connected
fuel cell (not shown). Further information on a reformed
20 hydrogen fuel system device of this type can be found in
U.S. Patent application, bearing attorney docket number
CT 00-013, simultaneously filed herewith, entitled
"HYDROGEN GENERATOR UTILIZING CERAMIC TECHNOLOGY",

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assigned to the same assignee and incorporated herein by this reference.

Referring now to FIG. 4, illustrated in simplified cross-sectional view is a thermal conductive structure, or via, generally referenced 30, such as that utilized in device 10 of FIGs. 1 and 2. Structure 30 is generally utilized for transfer of heat efficiently between chemical heater 28 and fuel vaporizer zone 16 and reaction zone 18, as well as between waste heat recovery 22 and reaction zone 18. As illustrated, thermal conductive structure 30 includes multi-layer ceramic layers 52 used in the fabrication of the monolithic structure 12. Identified is chemical heater section 28 and reaction zone chamber 18. Thermally conductive thick-film metal vias 54 thermally couple the top and bottom sections efficiently for good heat transfer. A thick-film metal layer 55 in intimate contact with chemical heater zone 28 and reaction zone 18 spreads and uniformly heats these sections.

Referring now to FIGs. 5 and 6, illustrated in simplified cross-sectional or plan view is a portion of fuel vaporizer 16. More particularly, illustrated in FIG. 5 is a serpentine channel 60, that is typically

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filled or coated with an inert porous ceramic material¹⁴ for thermal control. Alternatively, vaporizer 16 can be formed of a multiplicity of parallel channels 61, with an inert porous ceramic material for thermal control, in microfluidic communication with a porous diffuser 113. In each instance, serpentine channel 60 or parallel channel 61 includes a combined single liquid inlet feed 62 at one end and combining into a single vapor outlet at the other end 64. Serpentine channels 60 and parallel channels 61, are formed in the multi-layer ceramic structure, previously identified as 12. Reaction zone 18 includes similarly designed channels and features as found in the vaporizer zone, discussed above. More particularly, reaction zone 18 includes a reforming catalyst. The catalyst is present as channel wall coatings or as a porous packed bed of catalyst particles. One design goal is to reduce the dimensionality of the reactor channels to the order of a gas diffusion length and control gas residence times, or gas space velocity, that are favorable to reaction kinetics. A multiplicity of parallel reactor channels, generally similar to channels 61, provide for high gas throughput and minimize back pressure issues as related to channel

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blockage. It should be understood that it is anticipated by this disclosure that vaporizer zone 16 and reaction zone 18 may include in addition to, or in lieu of, serpentine channels 60 and parallel channels 61, any number of cavities or chambers, or any combination of channels, cavities, or chambers, thereof.

Illustrated in FIG. 7 in a simplified flow chart diagram, is the chemical reaction that takes place in device 10, and more particularly in reformer 14 of device 10. As illustrated, methanol 70 and water 72 are input into a steam reformer 74, generally similar to reformer 14 of FIGS. 1 and 2. Steam reformer 74 operates at a temperature of approximately 250°C to reform the input methanol 70 and water 72 into a reformed gas mixture, generally referred to as the hydrogen enriched gas. More particularly, in the presence of a catalyst, such as copper oxide, zinc oxide, or copper zinc oxide, the methanol 70 and water 72 solution is reformed into hydrogen, carbon dioxide, and some carbon monoxide. Steam reformer 74 operates in conjunction with an optional carbon monoxide cleanup 76, that in the presence of a preferential oxidation catalyst and air (or O₂), reforms a large percentage of the present carbon monoxide

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into carbon dioxide. This reformed gas mixture supplies
fuel through fuel output 78 to a fuel cell, generally
similar to fuel cell 32 of FIGs. 1 and 2.

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5 Illustrated in FIG. 8 is a schematic diagram of fuel
cell system 10 with integrated fuel reformer 14. As shown
in this diagram, fuel cell 32 can be operated directly
using output 22 from fuel reformer 14 without CO clean
up. In this instance fuel cell 32 is a high temperature
fuel cell. An advantage of the integration of a
traditional fuel cell stack 32 with the fuel processor
14, as illustrated, is the ability to operate fuel cell
32 at higher temperatures of 140-200°C by taking
advantage of the heat from reformer 14. High temperature
fuel cell membranes such as PBI (polybenzimidazole), or
15 similar polymer composition known as AEPBI can be used
in these applications. These fuel cell membranes
operating at temperatures greater than 120°C have much
higher tolerance up to a combined total of 5% (mol) CO
and methanol in the reformed hydrogen output from the
20 fuel processor. Use of these membranes eliminates the
need for a CO clean up step using the preferential
oxidation reactor zone and simplifies the system design.
In the alternative, by incorporating a CO preferential

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clean up reactor after steam reformer 14, a traditional
fuel cell 32 can be operated.

Accordingly, described is a fuel processor including
a ceramic carrier defining a reaction zone including a
catalyst. The ceramic carrier further includes a heat
source thermally coupled to the reaction zone chamber.
An inlet channel is supplied for input of the liquid fuel
to the fuel processor and an outlet channel is supplied
for the output of the reformed gas mixture. The fuel
processor device is formed as a monolithically integrated
structure, generally comprised of a plurality of thin
ceramic layers assembled and then sintered in such a way
as to provide for the closed heating zones in which the
encapsulated catalyst reforms the inlet fuel into mostly
hydrogen gas suitable for use with a fuel cell stack.

While we have shown and described specific
embodiments of the present invention, further
modifications and improvements will occur to those
skilled in the art. We desire it to be understood,
therefore, that this invention is not limited to the
particular forms shown and we intend in the appended
claims to cover all modifications that do not depart from
the spirit and scope of this invention.

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What is claimed is:

1. A fuel processor and integrated fuel cell
5 comprising:
a ceramic carrier defining a fuel reformer, the fuel
reformer including a reaction zone including a reforming
catalyst and a heat source thermally coupled to the
reaction zone;
10 an inlet channel for liquid fuel;
an outlet channel for hydrogen enriched gas; and
an integrated fuel cell, in microfluidic
communication with the outlet channel.
- 15 2. A fuel processor and integrated fuel cell as
claimed in claim 1 wherein the fuel reformer further
includes a vaporization zone.
- 20 3. A fuel processor and integrated fuel cell as
claimed in claim 1 wherein the heat source is an
integrated heater.

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4. A fuel processor and integrated fuel cell as claimed in claim 1 wherein the integrated heater is a resistive heater that is electrically driven.
- 5
5. A fuel processor and integrated fuel cell as claimed in claim 3 wherein the integrated heater is a chemical heater including a catalyst and arranged to oxidize fuel to produce heat.
- 10
6. A fuel processor and integrated fuel cell as claimed in claim 5 wherein the chemical heater further includes an air inlet for providing oxygen for the oxidation of the fuel and the inlet channel includes an opening to provide fuel to the chemical heater.
- 15
7. A fuel processor and integrated fuel cell as claimed in claim 3 wherein the integrated heater couples heat to the vaporization zone and reaction zone using thermally conductive structures.
- 20
8. A fuel processor and integrated fuel cell as claimed in claim 2 wherein one of the vaporization zone

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and the reaction zone include a plurality of parallel channels.

9. A fuel processor and integrated fuel cell as
5 claimed in claim 2 wherein one of the vaporization zone
and the reaction zone include a at least one serpentine
channel.

10. A fuel processor and integrated fuel cell as
10 claimed in claim 1 wherein the ceramic carrier is a
monolithic three-dimensional multilayer ceramic
structure.

11. A fuel processor and integrated fuel cell
15 comprising:

a monolithic three-dimensional multilayer ceramic
carrier structure defining a fuel reformer including a
vaporisation zone and a reaction zone including a
reforming catalyst, the ceramic carrier further including
20 an integrated heater thermally coupled to the reaction
zone;

an inlet channel for liquid fuel;

an outlet channel for hydrogen enriched gas; and

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an integrated fuel cell stack, in microfluidic communication with the outlet channel.

12. A fuel processor and integrated fuel cell comprising:

a monolithic three-dimensional multilayer ceramic carrier structure defining a fuel reformer including a vaporisation zone and a reaction zone including a reforming catalyst, at least one of the vaporization zone and the reaction zone including one of a plurality of parallel channels or at least one serpentine channel, the ceramic carrier further including an integrated heater thermally coupled to the vaporization zone and reaction zone using thermally conductive structures;

an inlet channel for liquid fuel, an outlet channel for hydrogen enriched gas, and an integrated fuel cell stack, in microfluidic communication with the outlet channel.

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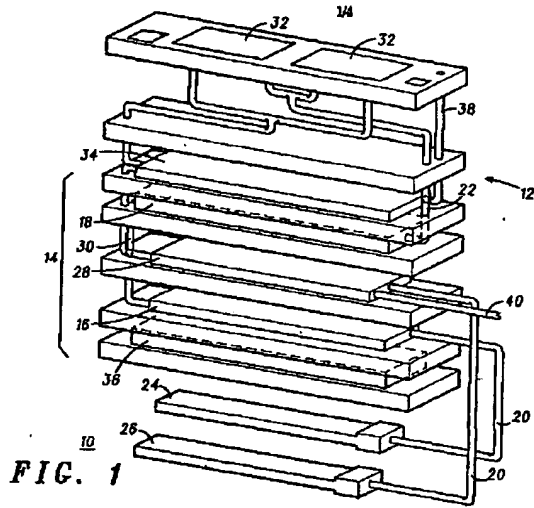
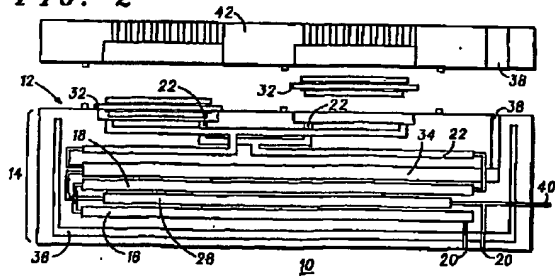


FIG. 1

FIG. 2



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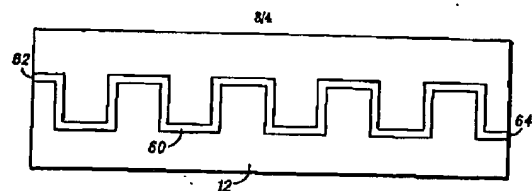


FIG. 5

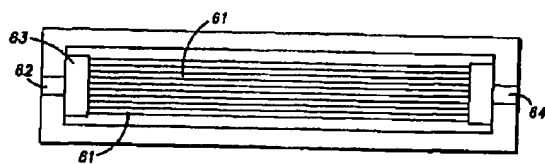


FIG. 6

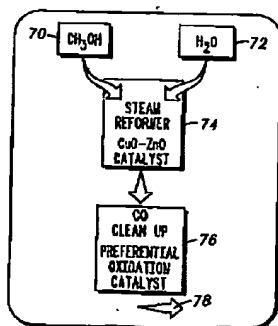


FIG. 7

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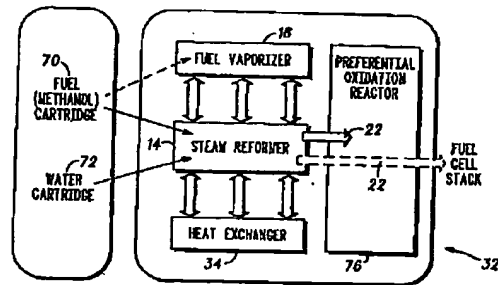


FIG. 8

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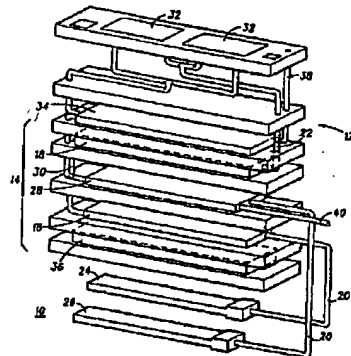
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(Continued on next page)

(54) Title: FUEL PROCESSOR WITH INTEGRATED FUEL CELL UTILIZING CERAMIC TECHNOLOGY



(57) Abstract: A fuel processor (10) and integrated fuel cell (12) including a monolithic three-dimensional multilayer ceramic carrier structure (14) defining a fuel reformer (16) and including an integrated fuel cell stack (22). The reformer includes a vaporization zone (18), a reaction zone (20) including a catalyst, and an integrated heater (24). The integrated heater is thermally coupled to the reaction zone. The fuel processor further includes an inlet channel (26) for liquid fuel and an outlet channel (72) for hydrogen enriched gas. The fuel processor is formed utilizing multi-layer ceramic technology in which thin ceramic layers are assembled that are sintered to provide miniature dimensions in which the monolithic catalyst converts or reforms fuel into a hydrogen enriched gas.

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【国際調査報告】

INTERNATIONAL SEARCH REPORT		Doc. no. Application No. PCT/US 02/26980
A. CLASSIFICATION OF SUBJECT MATTER IPC 7 H01M 4/00		
According to International Patent Classification (IPC) or to both national classification and IPC		
B. PRIOR ART Documents considered as prior art in the search IPC 7 H01M 4/00		
Documents considered as prior art in the search (not each document is included in the search)		
Documents which have been searched during the international search process of data base and, where practical, patent literature		
EPO-Internal, COMPENDEX, PAJ, MPI Data		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category	Details of document, with indication, where appropriate, of the relevant passages	Relevance to claims in:
X	DE 197 16 438 A (HEITZER JOERG DR) 22 October 1998 (1998-10-22) column 1, line 7 - line 11 column 1, line 40 - line 50 claims 1-3	1, 11, 12
P. X	EP 1 103 518 A (TOYOTA MOTOR CO LTD) 30 May 2001 (2001-05-30) paragraphs '00111, '00447, '00631, '00761, '00881, '00921 claims 1, 3, 5 figure 1	1-3, 11, 12
<input type="checkbox"/> Patent documents are based on the construction of the G. <input checked="" type="checkbox"/> Patent documents are based on the construction of the G.		
* Special categories of cited documents: (A) documents relating to the prior art of the invention as claimed in the claims of the patent application (B) documents relating to the prior art of the invention as claimed in the claims of the patent application (C) documents relating to the prior art of the invention as claimed in the claims of the patent application (D) documents relating to the prior art of the invention as claimed in the claims of the patent application (E) documents relating to the prior art of the invention as claimed in the claims of the patent application (F) documents relating to the prior art of the invention as claimed in the claims of the patent application (G) documents relating to the prior art of the invention as claimed in the claims of the patent application (H) documents relating to the prior art of the invention as claimed in the claims of the patent application (I) documents relating to the prior art of the invention as claimed in the claims of the patent application (J) documents relating to the prior art of the invention as claimed in the claims of the patent application (K) documents relating to the prior art of the invention as claimed in the claims of the patent application (L) documents relating to the prior art of the invention as claimed in the claims of the patent application (M) documents relating to the prior art of the invention as claimed in the claims of the patent application (N) documents relating to the prior art of the invention as claimed in the claims of the patent application (O) documents relating to the prior art of the invention as claimed in the claims of the patent application (P) documents relating to the prior art of the invention as claimed in the claims of the patent application (Q) documents relating to the prior art of the invention as claimed in the claims of the patent application (R) documents relating to the prior art of the invention as claimed in the claims of the patent application (S) documents relating to the prior art of the invention as claimed in the claims of the patent application (T) documents relating to the prior art of the invention as claimed in the claims of the patent application (U) documents relating to the prior art of the invention as claimed in the claims of the patent application (V) documents relating to the prior art of the invention as claimed in the claims of the patent application (W) documents relating to the prior art of the invention as claimed in the claims of the patent application (X) documents relating to the prior art of the invention as claimed in the claims of the patent application (Y) documents relating to the prior art of the invention as claimed in the claims of the patent application (Z) documents relating to the prior art of the invention as claimed in the claims of the patent application		
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